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16. ABSTRACT

Laboratory and field studies are underway to determine the effectiveness of activated carbon for removing volatile organic compounds from ground water. For fifteen C_1 through C_6 compounds* being considered for possible regulatory action, the adsorption isotherm capacity ranges from approximately 1 to 125 mg compound/gram activated carbon, at an equilibrium concentration of 500 µg/L. Because ground waters are typically low in background organic carbon, these adsorption isotherm data look promising for estimating adsorber performance. Additionally, physical and molecular properties of a contaminant may provide practical insight into its treatment by adsorption.

*tri- and tetrachloroethylene; cis- and trans-1,2-dichloroethylene; 1,1-dichloroethylene; methylene chloride; 1,1,1-trichloroethane; 1,2-dichloroethane; carbon tetrachloride; benzene; chlorobenzene; 1,2-, 1,3-, and 1,4-dichlorobenzene; and 1,2,4-trichlorobenzene

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REMOVAL OF VOLATILE ORGANIC CONTAMINANTS FROM GROUND WATER BY ADSORPTION

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Abstract

Laboratory and field studies are underway to determine the effectiveness of activated carbon for removing volatile organic compounds from ground water. For fifteen C_1 through C_6 compounds* being considered for possible regulatory action (1), the adsorption isotherm capacity ranges from approximately 1 to 125 mg compound/gram activated carbon, at an equilibrium concentration of 500 µg/L. Because ground waters are typically low in background organic carbon, these adsorption isotherm data look promising for estimating adsorber performance. Additionally, physical and molecular properties of a contaminant may provide practical insight into its treatment by adsorption.

Introduction

Lakes and rivers used as sources of drinking water typically contain 2 or more milligrams per liter (mg/L) of background organic carbon, but seldom exceed a few micrograms per liter (µg/L) of specific volatile organic compounds. Ground waters, on the other hand, generally are much lower in background organic carbon but some have been found to contain relatively high concentrations of volatile organics (1,2). Adsorption, long recognized as a treatment alternative for controlling problem organics in surface water, was a logical process to evaluate when the ground water contamination problem emerged. This report summarizes some of the laboratory and field projects conducted by the U.S. Environmental Protection Agency-Drinking Water Research Division (EPA-DWRD) on the topic of removing organic contaminants from ground water by adsorption.

Field Studies

The EPA-DWRD adsorption studies began in 1977 at two locations in New England. At each site, two small (4 cm diameter) columns, one containing granular activated carbon and the other a synthetic resin, were exposed to ground water containing industrial solvents. As Table 1 illustrates, work was subseqently expanded to include aeration with adsorption in New Jersey and New York, with the latter study incorporating steam regeneration of the adsorbents. The results from those studies have been published (3,4) but the following postscript is important: The synthetic resin used in these pilot scale

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*tri- and tetrachloroethylene; cis- and trans-1,2-dichloroethylene; 1,1-dichloroethylene; methylene chloride; 1,1,1-trichloroethane; 1,2-dichloroethane; carbon tetrachloride; benzene; chlorobenzene; 1,2-, 1,3-, and 1,4-dichlorobenzene; and 1,2,4-trichlorobenzene

projects, Ambersorb® XE-340, a product of the Rohm and Haas Company*, was an experimental material and although it had a relatively high adsorption capacity, the manufacturer does not plan to market the product. The future of full scale treatment by resin adsorption is therefore questionable.

As expected, the field studies showed a spectrum of adsorption effectiveness exists for the organic contaminants found in ground water. In general, the unsaturated hydrocarbons (those containing double bonds) were more effectively adsorbed than the saturated ones. Perhaps the most interesting finding from the field projects was the unexpectedly high adsorption capacities on granular activated carbon, relative to the capacities predicted by isotherm data published by Dobbs and Cohen (5) where pulverized activated carbon and two hours contact time were used to determine adsorption capacity. The actual capacities from the pilot scale field studies were 2 to 3 times higher than the isotherm prediction (3). This led to indepth laboratory work and the development of the adsorption capacity procedure explained in the next section.

An important note is the EPA-DWRD field studies were at sites where the ground water contained background organic carbon concentrations (measured as total organic carbon, TOC) less than 0.5 mg/L. Other studies (6,7) on ground water with TOC concentrations exceeding 1 mg/L have shown relatively lower adsorption capacities, thus underscoring the fact that competition for adsorption sites is a necessary consideration when generalizing on adsorption efficiency. Additionally, biological degradation of organic compounds during studies lasting several months may tend to increase the apparent capacity of activated carbon.

Laboratory Adsorption Studies

Certain criteria were established regarding the new adsorption capacity procedure that was developed because of poor correlation between isotherm predictions and actual field experiences. One, granular, rather than pulverized adsorbent was used, and two, a large reaction vessel (approximately 375L) was selected. The procedure has been named "macro-isotherm". A typical tank is shown in Figure 1. The granular activated carbon is external to the vessel in a Teflon® column, and a pump recirculates the water-adsorbate solution through the headspace free tank and column until "equilibrium"** is attained. Six vessels have been built. To the extent possible, construction materials are Teflon®, stainless steel and glass.

Details on the macro-isotherm procedure, its operation, and a comparison of adsorption capacities obtained by several methods are being prepared (8). Some of the results, however, are included in Figure 2, which is a summary of data from isotherm studies (5,9-11) for fifteen volatile organic compounds found in ground water. Also shown in Figure 2 is the mean adsorption capacity for each of these compounds at an equilibrium concentration of 500 µg/L. This adsorption capacity data, which ranges from approximately 1 to 125 mg

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**Equilibrium is defined as the condition where no statistically significant change in adsorbate concentration can be detected.

compound per gram of activated carbon, was correlated with molecular properties to aid in estimating adsorption behavior of newly identified organic compounds in ground water. An explanation of this preliminary work follows.

Molecular Properties and Adsorption Capacities

Just as Henry's Law constant is useful to compare the relative ease with which volatile organic compounds may be air stripped from water (12), the following question arises for adsorption: Are certain molecular properties useful for estimating an adsorption capacity? Earlier, Hofer and Manes (13) and Dobbs* suggested such an approach. Using the additive and constitutive properties shown in Table 2, and the mean adsorption capacity at an equilibrium concentration of 500 µg/L (Figure 2), the following observations were made.

Figure 3 shows a very scattered relationship between adsorption capacity and both molecular weight and density. Vapor pressure and solubility, on the other hand, (Figure 4) show a definite inverse relation to adsorption capacity. Although the scatter diagram is not shown, Henry's Law constant, which incorporates solubility and vapor pressure (and is useful for a priori aeration estimates) produced a very disappointing correlation with adsorption capacity.

The next properties examined were the parachor and the molar refraction. The parachor relates molar volume to surface tension and is defined by the equation:

$$P = \frac{M}{d} \gamma^{\frac{1}{4}} \qquad (14)$$

where:

P = parachor
M = molecular weight, g
d = density, g/mL
γ = surface tension, dynes/cm

The parachor of a compound can be estimated from the sum of the atomic and structural parachor equivalents (Table 3). When plotted against adsorption capacity (Figure 5), a general, direct relationship was found.

The molar refraction of a compound, which describes the relationship between molar volume and refractive index, is defined by the equation:

$$R_m = \frac{M}{d} \frac{n^2-1}{n^2+2} \qquad (14)$$

where:

Rm = the molar refraction, mL
M = molecular weight, g
d = density, g/mL
n = refractive index

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*Private communication, 1980.

Like the parachor, molar refraction can be approximated by adding the appropriate atomic and structural refraction equivalents shown in Table 4. The molar refraction similarly showed a general, direct relationship to adsorption capacity (Figure 5).

Certain molecular properties, therefore, may be useful for preliminary estimates of adsorption capacity when a "new" contaminant is discovered. In Figure 6, three arbitrary ranges of adsorption capacity (poor = <10 mg/g; fair = 10-50 mg/g; good = >50 mg/g) are shown with corresponding "fitted" curves ($y = bx^c$) for solubility, vapor pressure, molar refraction, and parachor. Assume, for example, a newly identified organic compound is known to have a low solubility and vapor pressure, and the parachor and molar refraction are calculated to be relatively high. That organic compound is likely to be well adsorbed on granular activated carbon. This approach is not intended to replace gathering the actual adsorption data, but it might be useful to obtain preliminary information for expected adsorption effectiveness where emperical data are lacking. The discretionary use of this approach cannot be over emphasized.

Discussion

The EPA-DWRD is continuing to evaluate adsorption as a process to remove organic compounds from ground water. The "macro-isotherm" procedure allows examination of adsorption rates concurrently with capacity, and upcoming studies will investigate the competitive effects between organic compounds and further define the influence of high background organic carbon concentrations. The technique may ultimately be useful for predicting full scale adsorber performance.

Approximately twelve water utilities in the United States are already using granular activated carbon to remove volatile organic contaminants from ground water. About one half of these utilities contract for this service from the activated carbon suppliers. This service is similar to that which has been available for several years to treat waste water. It consists of characterizing the water, installing the adsorption vessels, monitoring the effluent water quality, and replacing the spent activated carbon with virgin material when the concentration of a contaminant exceeds a predetermined effluent quality.

Adsorbers contain up to 9,000 kg (20,000 lbs) of activated carbon and are generally operated in series. Empty bed contact times ranging from 14 to 18 minutes are typical. Pretreatment by aeration, coagulation, and settling is being practiced at one water utility to remove iron and manganese and at least three water utilities are using aeration prior to adsorption to lengthen the service life of the activated carbon.

In some communities where a central treatment system is considered impractical, adsorption units containing activated carbon have been installed in homes having wells contaminanted with organic compounds (15). The question of providing proper maintenance to household units always arises. This aspect of evaluating home treatment units is being investigated jointly by the U.S. EPA and the National Sanitation Foundation (16).

With any treatment process, the effectiveness, and thus the overall cost, is dependent on a desired effluent quality. Currently (1982), no uniform agreement exists on what that quality should be, but it is

a topic undergoing critical review (1). Several water utilities, however, have elected to proceed with installing treatment to improve their water quality, and many of those utilities have selected adsorption.

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TABLE 1

ADSORPTION STUDIES* AT CONTAMINATED GROUND WATER SITES

Location	Years	Remarks
Connecticut	1977-1979	Pilot Scale: Granular activated carbon and synthetic resin (3)
New Hampshire	1977-1978	
New Jersey	1978-1980	Pilot Scale: Activated carbon and resin. Also aeration + activated carbon and resin (3)
New York	1978-1982	Pilot Scale: Activated carbon, resin with steam regeneration. (Also aeration) (4)

*U.S. Environmental Protection Agency, Drinking Water Research Division

TABLE 2

PHYSICAL AND CONSTITUTIVE PROPERTIES FOR SELECTED VOLATILE ORGANIC COMPOUNDS

	Chloroethylenes				
Parameter	tri-	tetra-	cis-1,2-di-	trans-1,2-di	1,1-di-
Molecular weight, g/mole	132	166	97	97	97
Density, g/mL	1.46	1.62	1.29	1.26	1.22
Solubility, mg/L (17,18)	1000	145	3500	6300	40
Vapor pressure, mm Hg (17,18)	74	19	206	271	495
Henry's Law Constant $x10^{-3}$ $\frac{atm\text{-}m^3}{mole}$ (19)	12	29	8*	5	150
Parachlor (14)	213	250	176	176	176
Molar refraction (14)	30	35	21	21	21

*Calculated from solubility and vapor pressure.

TABLE 2 (Cont'd)

PHYSICAL AND CONSTITUTIVE PROPERTIES FOR SELECTED VOLATILE ORGANIC COMPOUNDS

	Chloroethanes		Chloromethanes		Benzene	Chlorobenzenes				
Parameter	1,1,1-tri-	1,2-di-	carbon tetrachloride	methylene chloride		mono-	1,2-di-	1,3-di-	1,4-di-	1,2,4-tri-
Molecular weight, g/mole	133	99	154	85	78	113	147	147	147	182
Density, g/mL	1.34	1.24	1.59	1.33	0.89	1.11	1.31	1.29	1.25	1.45
Solubility, mg/L (17,18)	4950	8700	800	19,400	1780	448	100	123	79	30
Vapor pressure, mm Hg (17,18)	100	82	91	438	95	15	1	2	1	0.3
Henry's Law Constant $x10^{-3}$ $\frac{atm\text{-}m^3}{mole}$ (19)	5	1	30	3	6	4	2	2	2	1
Parachlor (14)	224	187	220	147	207	244	281	281	281	317
Molar refraction (14)	25	21	26	17	26	31	36	36	36	41

TABLE 3

ATOMIC AND STRUCTURAL PARACHOR EQUIVALANTS (14)

Carbon	4.8	Chorine	53.8	Double bond in C=C	23.2
Hydrogen	17.1	Bromine	68.0	Triple bond in C≡C	46.6
Oxygen	20.0	Iodine	91.0	Six-membered ring	6.1

TABLE 4

ATOMIC AND STRUCTURAL REFRACTION EQUIVALANTS (14)

Carbon	2.42 mL	Double bond in C=C	1.73 mL
Hydrogen	1.10	Triple bond in C≡C	2.40
Chlorine	5.97	Carbonyl oxygen	2.21
Bromine	8.87	Hydroxyl oxygen	1.53
Iodine	13.90	Ether oxygen	1.64

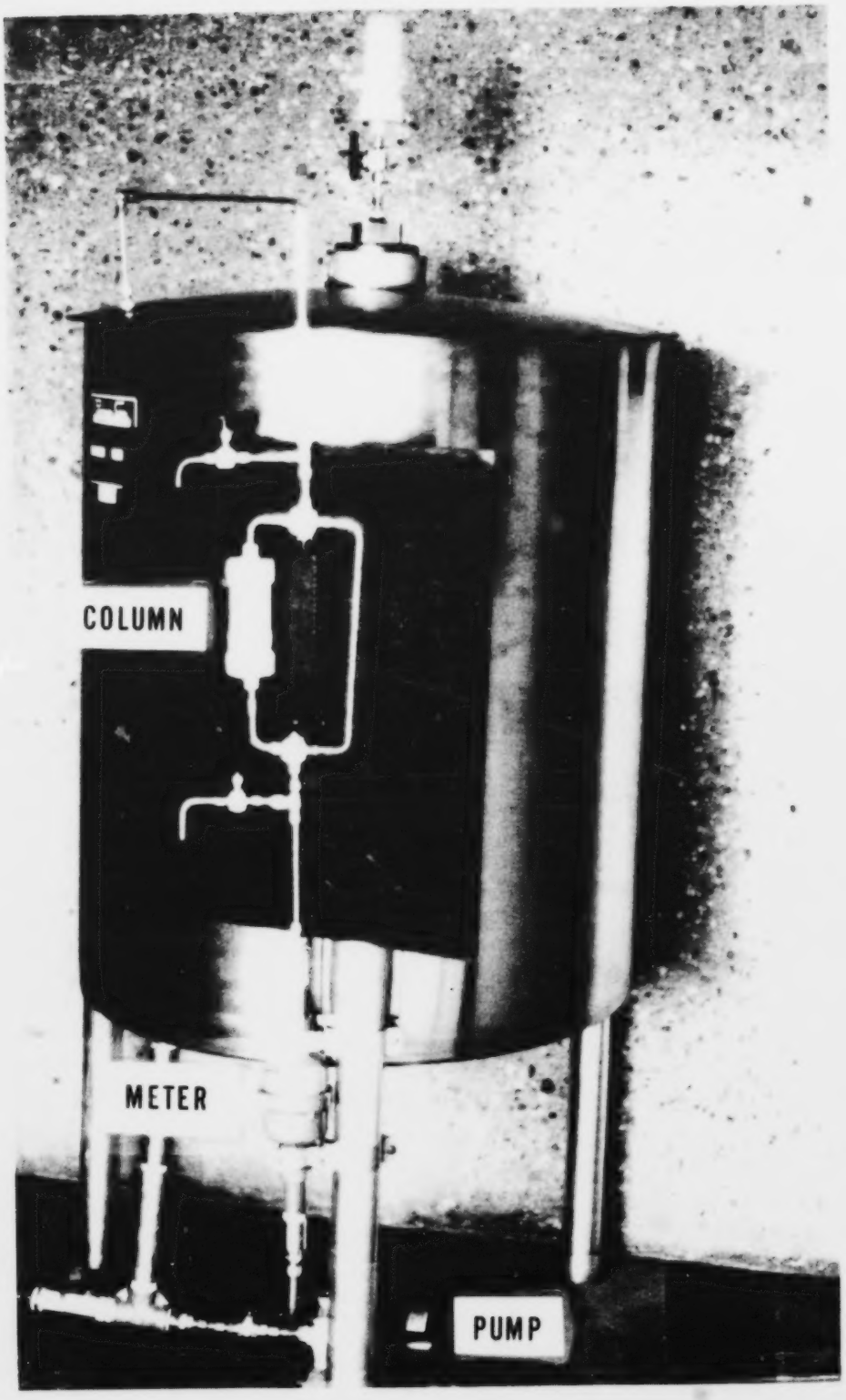
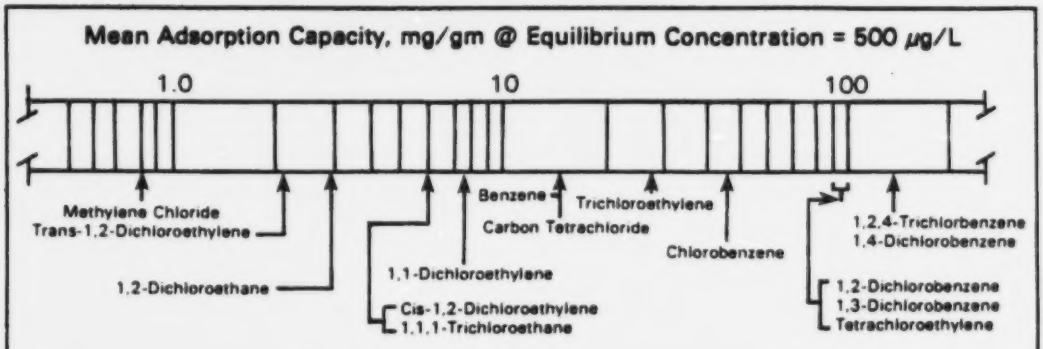


Figure 1. Vessel Used in Macro-isotherm Adsorption Studies



	Freundlich Parameters			Freundlich Parameters	
	K	1/n		K	1/n
Benzene	1.0	1.6*	trans-1,2-dichloroethylene	3.1	0.5*
	16.6	0.4**			
	49.3	0.6†	1,1-dichloroethylene	4.9	0.5*
	29.5	0.4††		16.0	0.5‡‡
	14.2	0.4§			
			Methylene chloride	1.3	1.2*
Carbon Tetrachloride	11.1	0.8*		1.6	0.7***
	28.5	0.8†			
	38.1	0.7**	Tetrachloroethylene	50.8	0.6*
	25.8	0.7††		84.1	0.4§§
	14.2	0.7§		273.0	0.6***
	14.8	0.4§§		128.0	0.5‡‡
	18.7	0.6‡‡			
Chlorobenzene	91.0	1.0*	1,2,4-trichlorobenzene	157.0	0.3*
1,2-dichlorobenzene	129.0	0.4*	1,1,1-trichloroethane	2.5	0.3*
				9.4	0.5§§
1,3-dichlorobenzene	118.0	0.4*		13.5	0.5‡‡
1,4-dichlorobenzene	121.0	0.5*	Trichloroethylene	28.0	0.6*
	226.0	0.4**		26.2	0.5‡
				28.2	0.4§§
1,2-dichloroethane	3.6	0.8*		63.1	0.5‡‡
	5.7	0.5§§			
	4.8	0.5‡‡			
cis-1,2-dichloroethylene	8.4	0.5§§			
	6.5	0.7‡			
	10.2	0.6‡‡			

Freundlich equation:
$x/m(mg/gm) = K\ C.(mg/l)^{1/n}$

*Filtrasorb® 300(5)
**Filtrasorb ® 400(9)
***Filtrasorb ® 400(10)
§Hydrodarco® 1030(9)
§§Witcarb® 950(11)
†Norit (9)
††Nuchar® WV-G(9)
‡Filtrasorb® 300(11)
‡‡Filtrasorb 400

Figure 2. Isotherm Adsorption Capacities on Activated Carbon

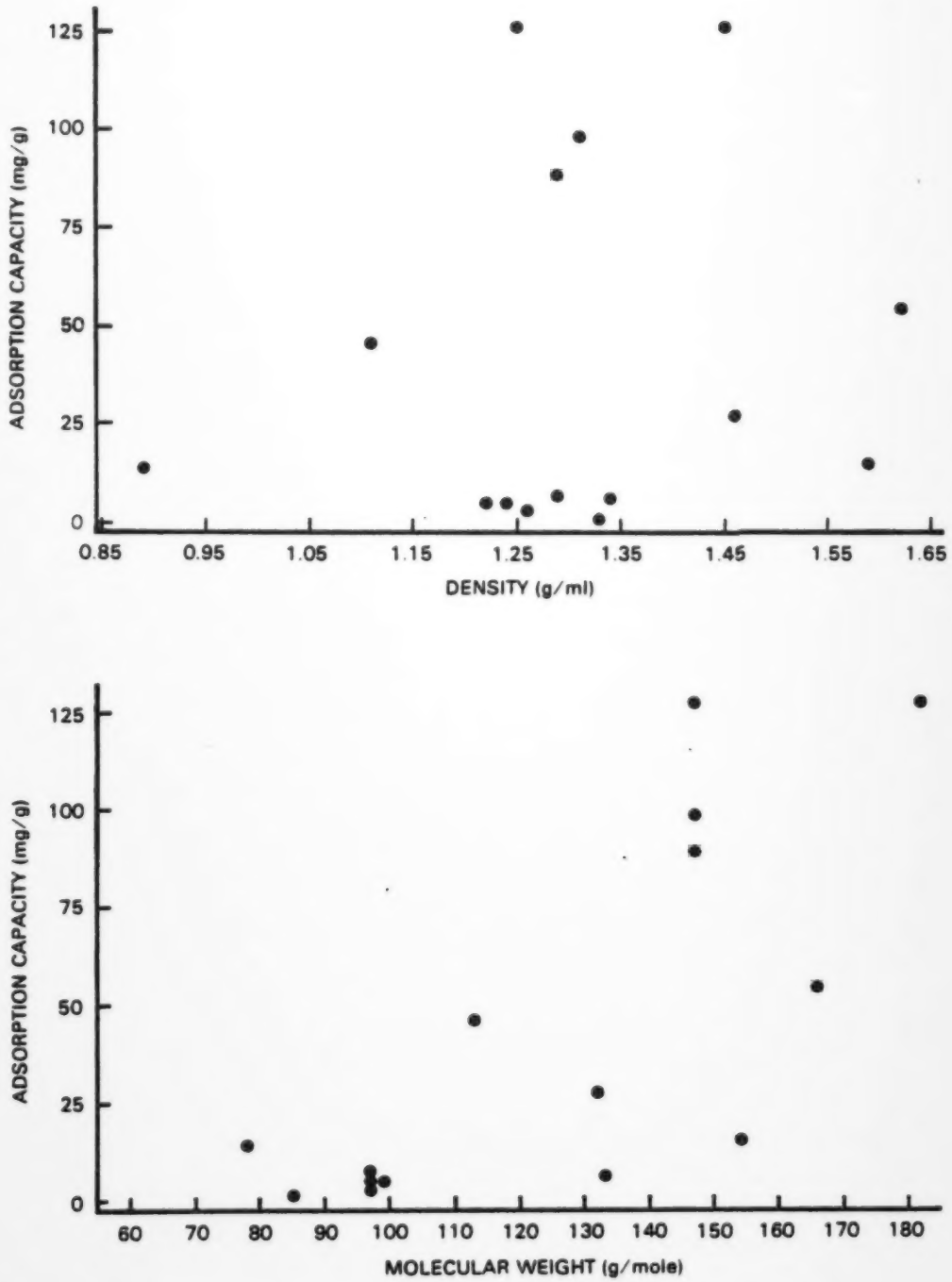
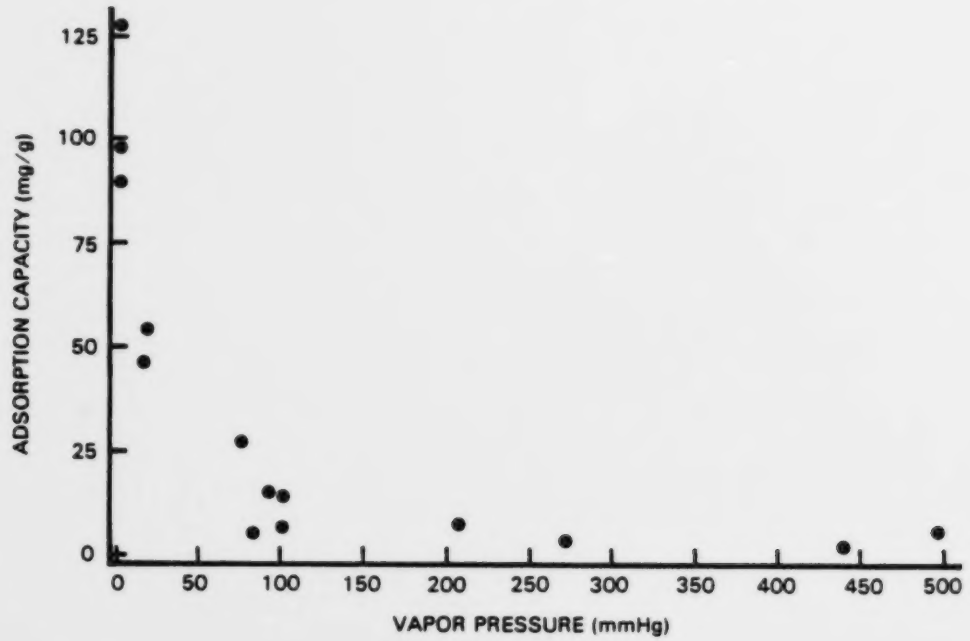


Figure 3. Comparison of Molecular Weight and Density to Adsorption Capacity (from Table 2 and Figure 2)



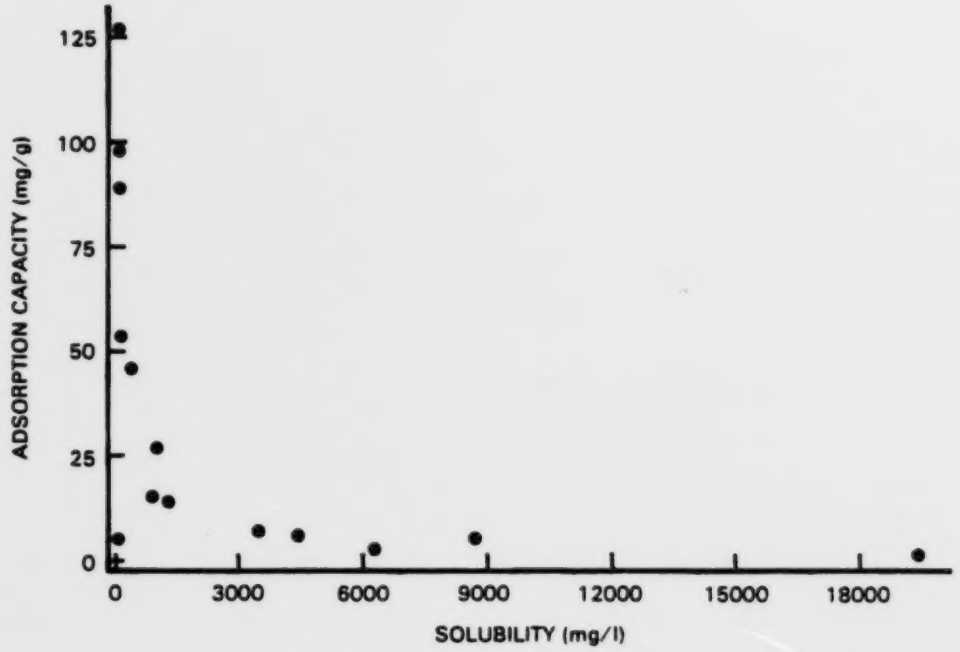


Figure 4. Comparison of Solubility and Vapor Pressure to Adsorption Capacity (from Table 2 and Figure 2)

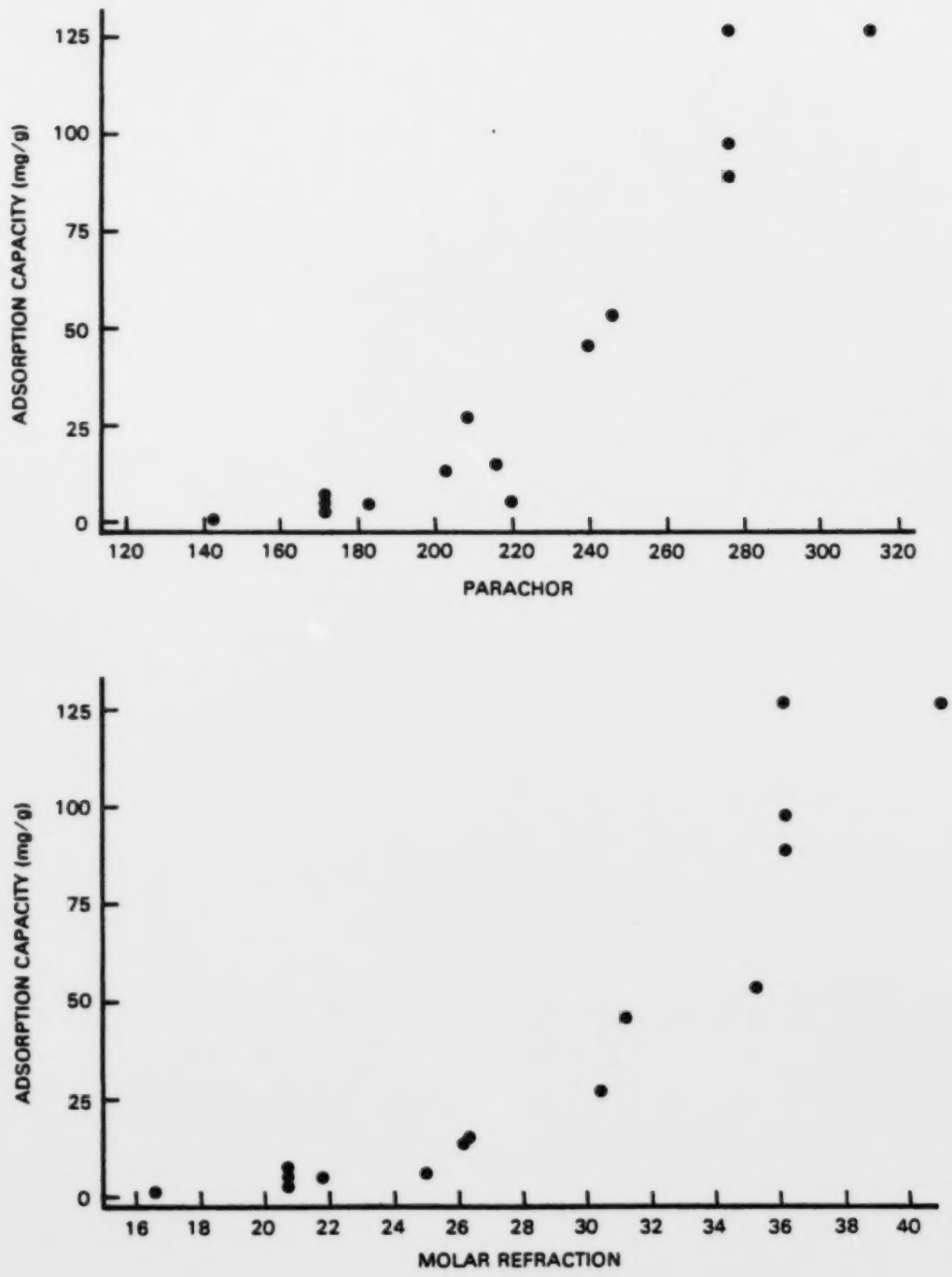


Figure 5. Comparison of Molar Refraction and Parachor to Adsorption Capacity (from Table 2 and Figure 2)

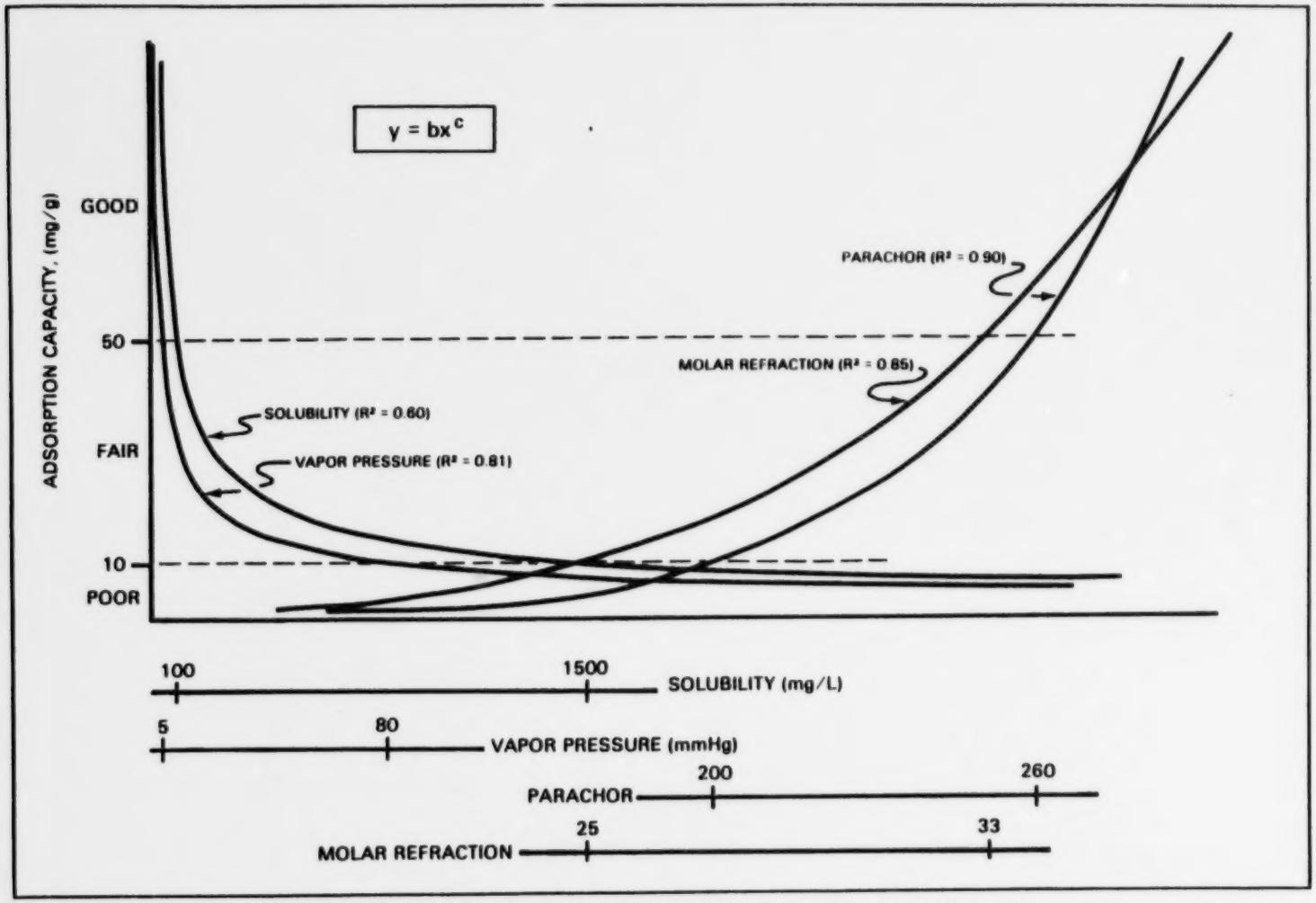


Figure 6. Use of Molecular Properties to Estimate Adsorption of Volatile Organic Compounds on Activated Carbon.